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Interactive comment on "Direct evidence for secondary ice formation at around –15 °C in mixed-phase clouds" *by* C. Mignani et al.

Anonymous Referee #2

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Authors present the experimental work where they collected the snow crystals, melted the crystals and visually observed the freezing of the crystal droplet. These results were used to understand more about the secondary ice formation and ice multiplication factors. These questions are challenging, and the community needs an understanding of these cloud processes for better representation in the cloud model. However, this study lacks appropriate experimental technique/methodology to answer these questions, and for this reason, the paper is not ready for the publication. I'm not sure if the major review could improve the paper further as substantial experimental work is involved.

There are a number of issues in the present experimental study. If no INP was observed within the crystal, it does not mean that crystal was formed through secondary

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ice formation mechanism. It is possible that a INP may have induced nucleation of ice, and still while INP is floating within the atmosphere may have detached from the ice crystal because the crystal evaporated or through some turbulent process. Now, this crystal when sampled had no INP. It is also possible that INP is present, but was deactivated while it went transformation (change in physical and chemical properties) during sampling, heating or droplet preparation. There are numerous studies in the literature that discusses the deactivation of INP. Such discussion is missing. Experiments are needed that investigate the ice nucleation efficiency of crystal melted droplets up to -37 degC (below this temperature homogeneous freezing is the dominant mode of ice nucleation) to understand more about the insoluble INPs, but for soluble INPs experiments should be investigated at homogeneous freezing temperatures too. Without such results, the conclusions regarding secondary ice formation cannot be inferred. Supporting experiments are needed to say why there was no INPs present (page 5 line 14). It would be just that the limitation of the experimental setup. In this study, the sample collection onto the cold stage is not done in clean air conditions. It is possible that crystals were contaminated with room air particles. Further, it is possible that these particles may have induced nucleation of ice but not the primary INP (the first INP that was responsible for freezing the droplet in the atmosphere before sampling). Without knowing the composition of residue it is difficult to infer which INP (primary or room air particulates) was responsible for freezing. It is not clear how section 2.3 supports the secondary ice formation analysis. Details such as validation and performance calibration of the cold stage (shown in Fig 1) under different temperature and humidity conditions are missing. Any results from previous studies who had attempted to study secondary ice formation should be shown in Figure 2 and 3. Discussion regarding nature of INP is missing. What are their composition and size?

One should use Ice-CVI (Mertes et al 2007) to sample only ice crystals, sublimate/evaporate these crystals, count the residues and investigate the ice nucleation propensity of a single residue. By comparing inlet ice crystal and residue concentrations one can infer some understanding regarding secondary ice formation.

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S. Mertes , B. Verheggen , S. Walter , P. Connolly , M. Ebert , J. Schneider , K. N. Bower , J. Cozic , S. Weinbruch , U. Baltensperger & E. Weingartner (2007) Counterflow Virtual Impactor Based Collection of Small Ice Particles in Mixed-Phase Clouds for the Physico-Chemical Characterization of Tropospheric Ice Nuclei: Sampler Description and First Case Study, Aerosol Science and Technology, 41:9, 848-864, DOI: 10.1080/02786820701501881.

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